

The Molecular Hubbard Hamiltonian: Field Regimes and Molecular Species

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The molecular Hubbard Hamiltonian (MHH) naturally arises for ultracold polar alkali dimer molecules in optical lattices. We show that, unlike ultracold atoms, different molecules display different many-body phases due to intrinsic variances in molecular structure. We also demonstrate a wide variety of experimental controls on molecules via external fields, including applied static electric and magnetic fields, an AC microwave field, and the polarization and strength of optical lattice beams. We provide explicit numerical calculations of the parameters of the MHH, including tunneling and direct and exchange dipole-dipole interaction energies, for the molecules ${}^6\text{Li}^{133}\text{Cs}$, ${}^{23}\text{Na}^{40}\text{K}$, ${}^{87}\text{Rb}^{133}\text{Cs}$, ${}^{40}\text{K}^{87}\text{Rb}$, and ${}^6\text{Li}^{23}\text{Na}$ in weak and strong applied electric fields. As case studies of many-body physics, we use infinite-size matrix product state methods to explore the quantum phase transitions from the superfluid phase to half-filled and third-filled crystalline phases in one dimension.

Molecules cooled to ultralow temperatures provide access to a wealth of exciting new physics, such as the study of chemical reactions at ultracold temperatures, precision tests of fundamental symmetries, novel means of quantum information processing, and many-body physics with many internal degrees of freedom [1]. The vast tunability proffered by molecules has led to many proposals to manipulate molecules with external fields in order to simulate outstanding unsolved quantum Hamiltonians motivated by solid state physics [2–4]. Taking a complementary approach, we address the immediately accessible many-body physics observable in upcoming experiments using ultracold molecules in optical lattices, in which external fields will not be fine-tuned. In this Letter, we use the molecular Hubbard Hamiltonian (MHH) to show that the first experiments with ultracold molecules in optical lattices will display a variety of many-body phases which are strongly dependent on molecular species and amenable to control with external fields.

For ultracold alkali atoms, interactions are well-modeled by a contact pseudopotential proportional to the s -wave scattering length, and the s -wave scattering length is readily tunable by using a Feshbach resonance. Hence, while the specific positions of Feshbach resonances may differ among different alkali species, essentially the same tunability of interactions is provided for all species. We demonstrate that the same is not true for ultracold molecules. Due to intrinsic variations in the structure of alkali dimer molecules, different molecules access different regimes of the MHH, and so display a variety of many-body phases. In this work, we take as examples the bosonic molecule ${}^{87}\text{Rb}^{133}\text{Cs}$ [5] and the fermionic molecules ${}^6\text{Li}^{133}\text{Cs}$ [6], ${}^{23}\text{Na}^{40}\text{K}$ [7], ${}^{40}\text{K}^{87}\text{Rb}$ [8], and ${}^6\text{Li}^{23}\text{Na}$ [9], all of which are being explored in current experiments. After identifying the MHH for a particular near-term experimental setup we discuss some of its experimental controls which are not available to ultracold atoms. We then provide explicit numerical values for the parameters appearing in a quasi one-dimensional

reduction of the MHH such as tunneling and dipolar interaction terms. To provide case studies of many-body physics, we numerically calculate quantum phase transitions from superfluid to crystalline phases at rational densities in one dimension using variational matrix product state techniques. By computing phase diagrams in terms of experimental parameters such as the strength of an applied electric field, we show that crystalline phases will not be seen for the molecules with the weakest dipole moments.

The molecular Hubbard Hamiltonian. The molecular Hubbard Hamiltonian (MHH) in the simplest experimental configuration is

$$\begin{aligned} \hat{H} = & - \sum_{ij\sigma;r_t} t_{j-i,\sigma} \hat{a}_{i,\sigma}^\dagger \hat{a}_{j,\sigma} - \sum_{\sigma\sigma'} \Omega_{\sigma\sigma'} \sum_i \hat{a}_{i,\sigma}^\dagger \hat{a}_{i,\sigma'} \\ & + \sum_{i\sigma} \Delta_\sigma \hat{n}_{i,\sigma} + \frac{1}{2} \sum_{ij\sigma\sigma';r_U} U_{j-i,\sigma\sigma'} \hat{n}_{i,\sigma} \hat{n}_{j,\sigma'} \\ & + \frac{1}{4} \sum_{ij\sigma_1\sigma_2\sigma'_1\sigma'_2;r_E} E_{j-i,\sigma_1\sigma_2\sigma'_1\sigma'_2} \hat{a}_{i,\sigma_1}^\dagger \hat{a}_{j,\sigma_2}^\dagger \hat{a}_{j,\sigma'_2} \hat{a}_{i,\sigma'_1}. \end{aligned} \quad (1)$$

Here σ labels the internal state of a molecule, and the operator $\hat{a}_{i,\sigma}$ ($\hat{a}_{i,\sigma}^\dagger$) represents either a hard-core bosonic or fermionic annihilation (creation) operator of a molecule in state σ on lattice site i . In order, the terms in Eq. (1) are tunneling of molecules between sites i and j with state-dependent tunneling energy $t_{j-i,\sigma}$, local transitions between molecular internal states $\sigma \rightarrow \sigma'$ driven by an AC microwave field \mathbf{E}_{AC} which has circular polarization in the lab frame and effective Rabi frequency $\Omega_{\sigma\sigma'}$, energetic detuning Δ_σ of a molecule in state σ from a reference ground state, *direct* dipole-dipole interactions $U_{j-i,\sigma\sigma'}$ between a molecule in state σ on site i and a molecule in state σ' on site j , and *exchange* dipole-dipole interactions $E_{j-i,\sigma_1\sigma_2\sigma'_1\sigma'_2}$ which exchange a quantum of rotation between molecules at sites i and j through the dipole-allowed transitions $\sigma'_1 \rightarrow \sigma_1$ and $\sigma'_2 \rightarrow \sigma_2$. The parameters r_t , r_U , and r_E represent cutoffs of the range of tunneling and direct and exchange dipole-dipole interactions, respectively, and set a consistent energetic order of approximation for the terms in Eq. (1) for a given set of field parameters. We note that these cutoffs generally

depend on the internal state, $r_t = r_{t,\sigma}$ etc., and on directionality, e.g. the range of tunneling will be shorter along directions in which the lattice potential is deeper.

The Hubbard parameters in Eq. (1) are defined as

$$t_{j-i,\sigma} \equiv - \int_{\text{BZ}} d\mathbf{q} e^{-i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)} E_{\sigma\mathbf{q}}/v_{\text{BZ}}, \quad (2)$$

$$\Delta_\sigma \equiv -t_{0,\sigma} - \omega\delta_{\bar{N},1}, \quad (3)$$

$$U_{j-i,\sigma\sigma'} \equiv (\mathcal{U}_{\sigma\sigma'\sigma'}^{0;ijji} + \mathcal{S}_{\sigma\sigma'}^{ijji}) \pm (\mathcal{U}_{\sigma\sigma'\sigma'}^{0;ijij} + \mathcal{S}_{\sigma\sigma'}^{ijij}), \quad (4)$$

$$E_{j-i,\sigma_1\sigma_2\sigma'_2\sigma'_1} \equiv \sum_{q=-1,1} [\mathcal{U}_{\sigma_1\sigma_2\sigma'_2\sigma'_1}^{q;ijji} \pm \mathcal{U}_{\sigma_1\sigma_2\sigma'_2\sigma'_1}^{q;ijij}], \quad (5)$$

$$\Omega_{\sigma\sigma'} \equiv E_{\text{AC}} d_{1,\sigma\sigma'}, \quad d_{q,\sigma\sigma'} \equiv \int d\mathbf{r} w_{0\sigma}^*(\mathbf{r}) \hat{d}_q w_{0\sigma'}(\mathbf{r}), \quad (6)$$

where v_{BZ} is the volume of the first Brillouin zone (BZ), \mathbf{R}_i is the coordinate of lattice site i , and the positive (negative) sign in Eqs. (4) and (5) refers to bosons (fermions). The Wannier functions $w_{i\sigma}(\mathbf{r})$ are the quasimomentum Fourier transforms of the lowest band Bloch functions

$$\hat{H}_{\text{sp}} \psi_{\sigma\mathbf{q}}(\mathbf{r}) = E_{\sigma\mathbf{q}} \psi_{\sigma\mathbf{q}}(\mathbf{r}), \quad (7)$$

which are the simultaneous eigenfunctions of the lattice translation operators and the single-particle Hamiltonian \hat{H}_{sp} . The dipole and short-range interaction integrals appearing in (4) and (5) are

$$\mathcal{U}_{\sigma_1\sigma_2\sigma'_2\sigma'_1}^{q;i_1i_2i'_2i'_1} = -2 \int d\mathbf{r} d\mathbf{r}' f_{\sigma_1\sigma'_1}^{i_1i'_1}(\mathbf{r}) \hat{d}_q \hat{d}_{-q} \frac{C_0^{(2)}(\mathbf{r}-\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|^3} f_{\sigma_2\sigma'_2}^{i_2i'_2}(\mathbf{r}'), \quad (8)$$

$$\mathcal{S}_{\sigma\sigma'}^{i_1i_2i'_2i'_1} = \int d\mathbf{r} d\mathbf{r}' f_{\sigma\sigma'}^{i_1i'_1}(\mathbf{r}) P(\bar{a}) f_{\sigma'\sigma'}^{i_2i'_2}(\mathbf{r}'), \quad (9)$$

where $f_{\sigma\sigma'}^{ii'}(\mathbf{r}) \equiv w_{i\sigma}^*(\mathbf{r}) w_{i'\sigma'}(\mathbf{r})$ is a Wannier function product, \hat{d}_q is the projection of the dipole operator along space-fixed spherical basis element \mathbf{e}_q , $C_0^{(2)}(\mathbf{R})$ is an unnormalized spherical harmonic, and $P(\bar{a})$ is a regularized s -(p)-wave pseudopotential [10] for bosons (fermions) which is assumed to have the universal dependence on the van der Waals length \bar{a} discussed in Ref. [11, 12]. We evaluate the integrals Eq. (8) using the ab initio method of Ref. [13]. As shown in that work, the dipole-dipole interaction parameters $U_{j-i,\sigma\sigma'}$ and $E_{j-i,\sigma_1\sigma_2\sigma'_2\sigma'_1}$ have a $\sim 1/|j-i|^p$ power-law tail with $p \gtrsim 3$ and a possibly significant exponentially decaying contribution when the lattice strengths are anisotropic.

The single particle Hamiltonian consists of kinetic, lattice, and internal components, and the internal contribution has been discussed in detail in past work [14]. We use the few-molecule parameters in Table I which have been calculated from density functional theory [15] or available experimental data [16]. The internal states of an alkali dimer molecule in the presence of a DC electric field $\mathbf{E}_{\text{DC}} = E_{\text{DC}} \mathbf{e}_z$ group into manifolds $|\bar{N}, M_N, \eta\rangle$. Here, \bar{N} denotes that the manifold adiabatically correlates with having N quanta of rotation as $E_{\text{DC}} \rightarrow 0$, M_N is the projection of rotation along \mathbf{e}_z , and η indexes the nuclear hyperfine degeneracy. Two such manifolds with $|M_N| \neq |M'_N|$ are well separated in energy provided that

TABLE I: The few-molecule parameters can differ by several orders of magnitude for experimentally relevant $^1\Sigma$ molecules [12, 15, 16]. In order, we give the permanent dipole moment d , the nuclear spins I_1 and I_2 , the rotational constant B_N , the nuclear quadrupole coupling constants $(eQq)_1$ and $(eQq)_2$, the nuclear g -factors g_1 and g_2 , the hyperfine spin couplings c_1 , c_2 , c_3 , and c_4 , the magneto-association field B , and the van der Waals length \bar{a} .

	$^6\text{Li}^{133}\text{Cs}$	$^{23}\text{Na}^{40}\text{K}$	$^{87}\text{Rb}^{133}\text{Cs}$	$^{40}\text{K}^{87}\text{Rb}$	$^6\text{Li}^{23}\text{Na}$
d/Debye	5.52	2.76	1.25	0.566	0.56
I_1	1	3/2	3/2	4	1
I_2	7/2	4	7/2	3/2	3/2
B_N/GHz	6.5202	2.8268	0.504	1.114	12.7355
$(eQq)_1/\text{MHz}$	3×10^{-4}	-0.167	-0.872	0.45	8×10^{-4}
$(eQq)_2/\text{MHz}$	0.187	0.796	0.051	-1.41	0.687
g_1	0.822	1.479	1.834	-0.324	0.822
g_2	0.738	-0.324	0.738	1.834	1.479
c_1/Hz	15.2	110.6	98.4	-24.1	83.2
c_2/Hz	3476.0	-90.9	194.1	420.1	802.2
c_3/Hz	53.1	-46.5	192.4	-48.2	196.2
c_4/Hz	620.8	-466.2	17345.4	-2030.4	212.3
B/Gauss	843.5	139.7	181.7	545.9	745
\bar{a}/nm	25	23.5	23.5	6	2.5

the scaled DC field strength $\beta_{\text{DC}} \equiv dE_{\text{DC}}/B_N \gtrsim 0.2$. In addition to \mathbf{E}_{DC} , we consider an applied DC magnetic field $\mathbf{B} = B\mathbf{e}_z$. The values of B we use, collected in Table I, correspond to the positions of the Feshbach resonances used for magneto-association [5–9]; these are the magnetic fields naturally and easily used in experiments. The Zeeman splittings and intrinsic couplings between hyperfine sublevels produce intra-manifold splittings which are typically a few tens to hundreds of kHz.

In order to induce rotational transitions between states in manifolds $|\bar{0}, 0, \eta\rangle$ and $|\bar{1}, \pm 1, \eta'\rangle$, we require a single AC microwave field with frequency ω and circular polarization with respect to the DC field axis; such fields are already used in experiments [16]. The detuning Δ_σ denotes how far from a single-molecule resonance state σ is, and the effective Rabi frequency $\Omega_{\sigma\sigma'}$ expresses the strength of transitions from state σ to state σ' . In strong B fields where the intra-manifold splitting is typically larger than e.g. the dipole-dipole interaction energy, the AC field determines which states are dynamically accessible. Hence, the number of states involved in the MHH dynamics can be modified by the static field configuration and the frequency and power of the AC field, resulting in a tunable quantum complex system [14]. The number of dynamically accessible internal states can be quite sizable, typically a few tens, due to a large hyperfine degeneracy, see I_1 and I_2 in Table I.

Finally, we consider that the molecules are subject to a simple cubic optical lattice potential consisting of

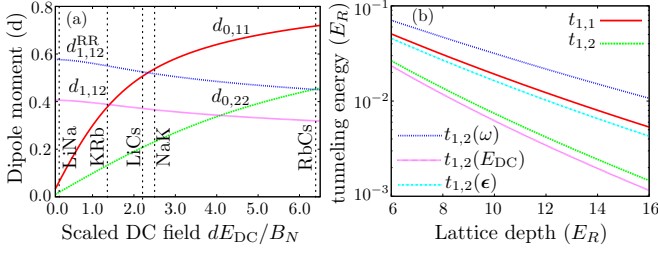


FIG. 1: (Color online) *Examples of experimental controls not available to atoms.* (a) An applied electric field E_{DC} gives rise to single-particle states with tunable expected dipole moments. Vertical dashed lines indicate the position of $E_{DC} = 5 \text{ kV/cm}$ for the various molecules. (b) The difference in tunneling energy between internal states may be tuned by a variety of controls which have no counterparts for atoms such as the DC field strength E_{DC} , the angle between the lattice polarization ϵ and \mathbf{E}_{DC} , and the frequency ω of the microwave field.

three pairs of counter-propagating laser beams, $V_{\text{latt}}(\mathbf{r}) = \sum_{\eta \in \{x,y,z\}} E_{\eta}^2 \cos^2(\pi\eta/a)\epsilon_{\eta}^* \cdot \tilde{\alpha} \cdot \epsilon_{\eta}$. Here, $\tilde{\alpha}$ is the polarizability tensor of the molecules, E_{η}^2 is the intensity of the laser field along direction η , ϵ_{η} is the polarization of the field along η , and $a = 545 \text{ nm}$ is the lattice spacing. In the numerical examples we choose the polarizations $\epsilon_x = \mathbf{e}_y$, $\epsilon_y = \mathbf{e}_z = \mathbf{e}_x$ corresponding to current experiments [17]. The ratio of the components of the polarizability due to coupling to Σ and Π states at the given wavelength, α^0/α^1 , is taken to be 0.223 [18]. Due to the fact that the molecular polarizability tensor is rank two, non- \mathbf{e}_z polarized light couples together the states in the manifold $|\bar{1}, \pm 1, \eta\rangle$ [19] and the tunneling amplitude $t_{p,\sigma}$ depends on the internal state and the polarization, see Table II and Fig. 1(b). Also as a result of the coupling in the manifold $|\bar{1}, \pm 1, \eta\rangle$, the single-particle eigenstates are in general a quasimomentum-dependent superposition of the states which diagonalize the internal Hamiltonian in free space, and so the dipole moments generally cannot be taken outside the integration in Eq. (8).

Tunability of MHH parameters for two internal states. The given experimental setup provides several controls which have no counterpart in atoms. To provide explicit examples of this tunability, we now focus on a special case of the MHH in which only two internal states are populated, $\sigma \in \{1, 2\}$, and the lattice is strongly confining in the y and z directions. The condition of only two internal states is well-realized for current experiments with large B fields and weak microwave power [8, 16]. The state $|\sigma = 1\rangle$ is taken to be the absolute ground state, and the state $|\sigma = 2\rangle$ is the lowest energy eigenstate which maximizes the \hat{d}_1 transition dipole moment with $|\sigma = 1\rangle$ [20].

Figure 1(a) shows the dependence of the dipole moments $d_{q,\sigma\sigma'}$, see Eq. (6), on the scaled DC field strength β_{DC} . The resonant moments $\{d_{0,\sigma\sigma'}\}$, which are the expected dipole moments of the states $\{|\sigma\rangle\}$, give rise to the direct dipole-dipole interactions $U_{j-i,\sigma\sigma'}$, and

TABLE II: Order of magnitude differences in Hubbard parameters give rise to a variety of phases. Here we collect the many-body parameters of the MHH Eq. (1) for $V_y = V_z = 55E_R$, $V_x = 8E_R$, the few-molecule parameters given in Table I, and the E_{DC} and species given in the table. The value $E_{DC} = 5 \text{ kV/cm}$ is a typical strong field in experiments. We do not include Ω , as it is widely tunable by adjusting the power of the applied AC microwave field.

	$^6\text{Li}^{133}\text{Cs}$	$^{23}\text{Na}^{40}\text{K}$	$^{87}\text{Rb}^{133}\text{Cs}$	$^{40}\text{K}^{87}\text{Rb}$	$^6\text{Li}^{23}\text{Na}$
$E_{DC} = 1 \text{ kV/cm}$					
$U_{1,11}$ (kHz)	0.70	0.218	0.253	0.002	2×10^{-5}
$U_{1,22}/U_{1,11}$	0.084	0.085	0.108	0.082	0.08
$U_{1,12}/U_{1,11}$	0.28	0.29	0.32	0.28	0.28
$t_{1,1}/U_{1,11}$	0.33	2.36	0.58	93.74	5×10^5
$t_{1,2}/U_{1,22}$	1.76	12.37	2.25	508.9	3×10^6
$E_{1,1212}/U_{1,11}$	7.93	6.05	0.99	20.97	2×10^3
$E_{DC} = 5 \text{ kV/cm}$					
$U_{1,11}$ (kHz)	8.97	2.48	0.970	0.049	5×10^{-4}
$U_{22}^1/U_{1,11}$	0.14	0.16	0.35	0.11	0.08
$U_{12}^1/U_{1,11}$	0.38	0.40	0.59	0.32	0.28
$t_{1,1}/U_{1,11}$	0.026	0.20	0.15	5.13	2×10^3
$t_{1,2}/U_{1,22}$	0.068	0.48	0.14	19.75	1.2×10^5
$E_{1,1212}/U_{1,11}$	0.519	0.43	0.18	1.05	114

the transition moments $\{d_{1,\sigma\sigma'}\}$, which give the strength of a dipole-allowed transition $\sigma \rightarrow \sigma'$, yield the exchange terms $E_{j-i,\sigma_1\sigma_2\sigma'_1\sigma'_2}$. The transition moments are strongly affected by hyperfine structure and lattice polarization effects as shown by the discrepancy between $d_{1,12}$ and $d_{1,12}^{RR}$, the latter being computed in the absence of a lattice. In Fig. 1(b) we display the tunneling energies of two internal states as a function of the quasi-1D lattice depth $V_x \equiv \langle \sigma = 1 | E_x^2 \epsilon_x^* \cdot \tilde{\alpha} \cdot \epsilon_x | \sigma = 1 \rangle$ measured in units of the recoil energy $E_R = \hbar^2 \pi^2 / 2ma^2$. The disparity between the tunneling in different internal states can be altered by changing the strength of the DC field, tilting the lattice polarization, or changing the frequency of the AC field, as indicated by the various curves.

Many-body case studies. To demonstrate the emergent many-body features of Eq. (1) in near-term experimental setups, we present many-body case studies employing the infinite-size variational ground state search using matrix product states (iMPS) for 1D systems [21]. We choose the energetic cutoff defining r_t etc. in Eq. (1) to be 1 Hz, and the field configurations are chosen such that terms not included in Eq. (1) such as assisted tunneling and tunneling outside of the quasi-1D domain fall below this threshold. These cutoffs account for the fact that many-body phases induced by long-range interactions may be fragile against other terms in the Hamiltonian, non-conservative effects such as heating from the optical lattice, and lifetime constraints of the ultracold gas.

We begin by considering the phase diagram of Eq. (1)

in a quasi-1D geometry and absent an AC field. Without the AC field only the absolute ground state $\sigma = 1$ is populated, and so the MHH reduces to

$$\hat{H} = -\sum_{ij;r_t} t_{j-i} \hat{a}_i^\dagger \hat{a}_j + \frac{1}{2} \sum_{ij;r_U} U_{j-i} \hat{n}_i \hat{n}_j. \quad (10)$$

At half filling, $\rho = 1/2$, a phase transition occurs at strong coupling (small t_1/U_1) to a crystalline phase (CP) with long-range order in the density correlation function $\mathcal{N}(r) \equiv \langle \Delta \hat{n}_0 \Delta \hat{n}_r \rangle$, $\Delta \hat{n}_i = \hat{n}_i - \rho$; an exponential decay of the single-particle density matrix $\mathcal{A}(r) \equiv \langle \hat{a}_0^\dagger \hat{a}_r \rangle$; and a non-vanishing single-particle gap corresponding to fixed density for a range of chemical potential μ , see Fig. 2. To within numerical resolution, we find that the CP melts directly into a superfluid with algebraic decay of both $\mathcal{N}(r)$ and $\mathcal{A}(r)$ and no single-particle gap, and there is no region of supersolid phase displaying simultaneous long-range order in $\mathcal{N}(r)$ and algebraic decay of $\mathcal{A}(r)$. Due to their small dipole moment, $^{40}\text{K}^{87}\text{Rb}$ and $^6\text{Li}^{23}\text{Na}$ do not display the CP phase transition for fields $E_{\text{DC}} \lesssim 5\text{ kV/cm}$, and so are not included in Fig. 2. The absence of the CP transition for these molecules is an example of how different molecules display different many-body physics.

When the interaction U_x is a convex function of x , it has been conjectured that regions of rational density other than $\rho = 1/2$ can occupy finite regions in the phase diagram [22]. To study the accessibility of such phases, we investigate the CP at filling $\rho = 1/3$ for $^6\text{Li}^{133}\text{Cs}$ in Fig. 2(f). Here we fix E_{DC} and vary the quasi-1D lattice height V_x in order to change the relative strengths of interaction and tunneling. While it is possible to achieve this phase with the other molecules, the equilibration timescales will be longer than the lifetime of typical experiments due to the smaller dipole interaction energy which sets the timescale, see Table II. Hence, $^6\text{Li}^{133}\text{Cs}$ is the best choice for investigating phases due to strong direct interactions. Similarly, other CPs with densities $\rho < 1/3$ could be seen in principle, but the requirement of very weak tunneling with respect to interactions is suggestive that such configurations may be difficult to equilibrate on reasonable timescales as well as obtaining a sufficiently cold system. We note that the $\rho = 1/3$ CP is unique to molecules with long-range dipole-dipole interactions; there is no analog of this phase in atoms with short-range interactions.

The simplest situation utilizing the internal structure of the molecule is when there is exactly one molecule per lattice site and only two internal states are accessible. Due to the hard-core constraint the translational degrees of freedom are frozen out and only the internal degrees of freedom are dynamical. When the two internal states of the molecule are mapped to an effective two-component spin via the standard transformation $\hat{S}_i^z \equiv (\hat{n}_{1i} - \hat{n}_{2i})/2$, $\hat{S}_i^+ \equiv \hat{a}_{1i}^\dagger \hat{a}_{2i}$, $\hat{S}_i^- = (\hat{S}_i^+)^\dagger$, the MHH for either bosons or fermions maps, up to constant terms, onto a long-range

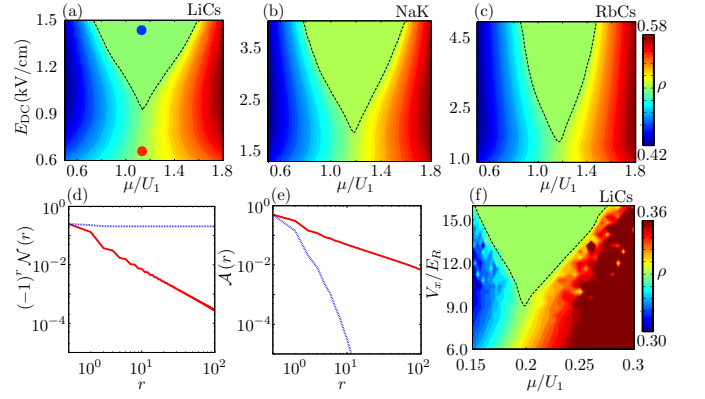


FIG. 2: (Color online) *Phase diagram of the MHH in the absence of an AC field.* (a) Density phase diagram for LiCs, showing the $\rho = 1/2$ crystalline phase (CP) with non-vanishing single-particle gap. The dashed line is a guide to the eye for the region of fixed $\rho = 1/2$. (b, c) analogs of (a) for NaK and RbCs, respectively. (d) Density correlation function $\mathcal{N}(r)$ with period-two oscillation removed for the points marked in (a). (e) Single-particle density matrix $\mathcal{A}(r)$ for the points marked in (a). (f) Density phase diagram for LiCs near the $\rho = 1/3$ CP transition at fixed $E_{\text{DC}} = 3.5\text{ kV/cm}$ showing small density fluctuations induced by our use of the grand canonical ensemble [21].

XXZ model in transverse and longitudinal fields

$$\hat{H} = \frac{1}{2} \sum_{ij;r_X} X_{j-i} \hat{S}_i^+ \hat{S}_j^- - 2\Omega \sum_i \hat{S}_i^x + h_z \sum_i \hat{S}_i^z \quad (11) \\ + \frac{1}{2} \sum_{ij;r_I} I_{j-i} \hat{S}_i^z \hat{S}_j^z.$$

Here, $I_{j-i} \equiv U_{j-i,11} - 2U_{j-i,12} + U_{j-i,22}$, $X_{j-i} \equiv E_{j-i,1212}$, and $h_z \equiv (\Delta_1 - \Delta_2) + \sum_{j-i;r_U} (U_{j-i,11} - U_{j-i,22})$. For the given experimental setup, both the Ising-type coupling I_{j-i} and the XX-type coupling X_{j-i} are positive, corresponding to antiferromagnetic interactions. This model has been studied in 1D recently without the Ω term [23]. As Ω is increased relative to the other terms in the Hamiltonian, the system transitions to a trivial paramagnetic phase with nonzero magnetization along x , corresponding to strong on-site correlation of the internal state. We stress that the ratios of the various spin couplings depend in an essential way on the molecular species, see Table II.

We have presented the molecular Hubbard Hamiltonian, the natural Hamiltonian governing the many-body physics of ultracold molecules in optical lattices. Molecules are sensitive to experimental controls which are not relevant for atoms such as an applied DC electric field, the polarization of optical lattice light, and the frequency and power of an AC microwave field driving rotational transitions. The response of a molecule to external fields is also much more dependent on molecular species than is the case for atoms. Our results establish that the molecules employed in current experiments will display a range of species-dependent many-body features.

We acknowledge useful discussions with J. Aldegunde, J. L. Bohn, A. V. Gorshkov, J. Hutson, K. Maeda,

and M. Weidemüller. This work was supported by the Alexander von Humboldt Foundation, the Heidelberg Center for Quantum Dynamics, AFOSR, NSF, and GECO.

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Supplementary material to “The Molecular Hubbard Hamiltonian: Field Regimes and Molecular Species”

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INFINITE-SIZE VARIATIONAL GROUND STATE SEARCH USING MATRIX PRODUCT STATES

Infinite-size variational ground state search using matrix product states (iMPS) is a numerical method which finds a representation for the ground state of an infinite one-dimensional lattice model [1]. The state is assumed to be invariant under shifts by a user-specified number of lattice sites q , and hence can be represented in terms of a periodically repeating unit cell with q lattice sites. The unit cell is decomposed as a matrix product state (MPS) with finite entanglement cutoff χ measured by the Schmidt rank [2], and this unit cell is iteratively optimized via the solution of an eigenvalue problem. The optimization reaches an end when the density matrix $\hat{\rho}$ obtained by tracing over all sites to the right of a given point in the infinite lattice changes by less than a tolerance ϵ between successive iterations. We measure that change via the *orthogonality fidelity*

$$F_{\text{ortho}} \equiv \text{Tr} \sqrt{\sqrt{\hat{\rho}_n} \hat{\rho}_{n-1} \sqrt{\hat{\rho}_n}}, \quad (12)$$

where the subscript on $\hat{\rho}$ denotes the optimization cycle index. In practice, we choose the orthogonality fidelity tolerance to be $\epsilon = \bar{\epsilon}_{\text{local}}$, where $\bar{\epsilon}_{\text{local}}$ is unit-cell averaged 2-norm distance between the optimal unit cell and its representation as an MPS with fixed entanglement cutoff χ . That is, deviations of the density matrix $\hat{\rho}$ between successive iterations are only from the errors due to a finite cutoff of entanglement. Once the unit cell has converged, a standard orthonormalization procedure is used to define a normalized state which conforms to a known canonical MPS form on the infinite lattice [3]. From this state, any desired observable may be computed.

After optimizations have been performed with a range of entanglement cutoffs χ , the effects of finite entanglement cutoff can be discerned by a scaling analysis in χ . It is known rigorously that MPSs with a finite cutoff χ cannot display power-law decay of correlation functions for arbitrary distances. Hence, for a given χ , there exists a distance r_χ beyond which a correlation function with a power-law decay begins to decay exponentially, and r_χ is an increasing function of χ . In addition, the scaling for some universal quantities, such as the correlation length, have a known scaling relation with χ which depends only on the data of the conformal field theory describing criticality in the infrared limit [4]. An example of the scaling of correlation functions with χ is given in Fig. 3, which shows the scaling of the single-particle density matrix $\mathcal{A}(r)$ in the superfluid and crystalline phases of Eq. (10) near half filling. All results given in the main text have been checked for convergence in χ and q , the unit cell length.

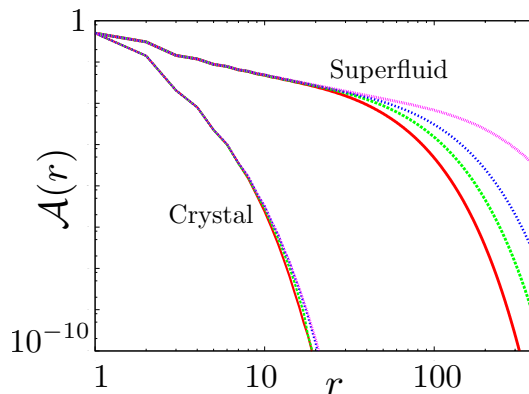


FIG. 3: *Scaling of single-particle density matrix with entanglement cutoff in iMPS.* The single-particle density matrix $\mathcal{A}(r)$ computed by iMPS for $\chi = 12$ (red), 16 (green), 20 (blue), and 40 (purple) in the crystalline and superfluid phases. In the crystalline phase, $\mathcal{A}(r)$ decays exponentially and so the results converge for finite χ . In the superfluid phase $\mathcal{A}(r)$ has an algebraic decay, and the range over which this decay is well-approximated increases with χ .

Long-range interactions are accommodated by representing the Hamiltonian as a matrix product operator (MPO) [5]. When the interaction cutoff r_U is less than 6, we use a finite-range interaction on r_U sites, but for larger cutoffs we use an infinite-range Hamiltonian obtained by fitting the function U_p to a series of exponentials [6].

We do this out of numerical convenience; the infinite-range decomposition requires fewer numerical resources for the same level of accuracy when $r_U > 6$.

The density plot near the $\rho = 1/3$ crystalline phase, Fig. 2(f) of the main text, appears noisy. This apparent noise is an artifact of having a finite resolution of points in the plot and using the grand canonical ensemble. We demonstrate this point in Fig. 4. Panel (a) displays the density near the phase transition, and panel (b) shows the same plot except that the region to the left of the solid vertical line has been computed with double the resolution in chemical potential. The density fluctuation spots become smaller for higher resolution, and are concentrated on the border between two regions of fixed density. Hence, as the resolution increases, the effect of these fluctuations will become less prominent. Density fluctuations on the border between superfluid regions of fixed density are also seen for finite systems in the grand canonical ensemble, but do not appear in the canonical ensemble [7]. Finite-size effects introduce a non-vanishing single-particle gap, and this inherently limits the resolution of the chemical potential for the grand canonical ensemble in a finite-volume system.

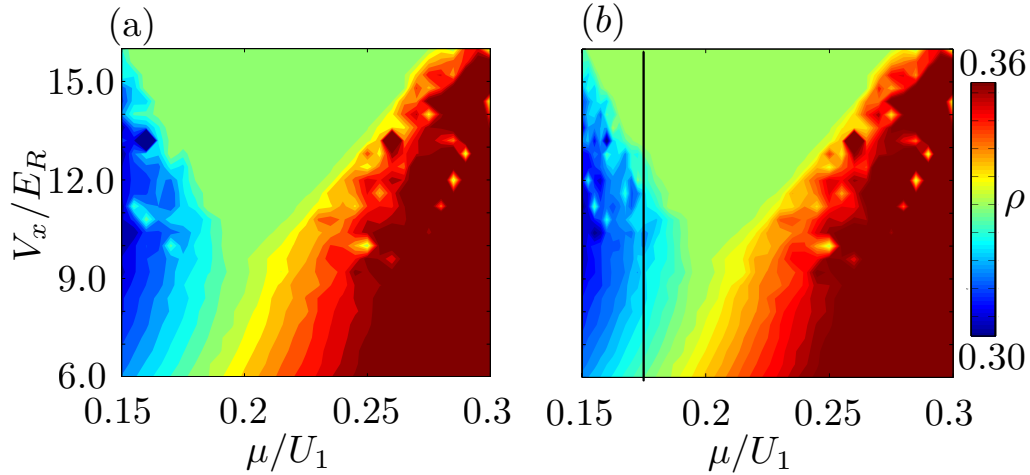


FIG. 4: *Number fluctuations in the grand canonical ensemble.* (a) Density plot of the region near $\rho = 1/3$. (b) Same as in (a), but the region to the left of the solid vertical line is computed with twice the resolution in chemical potential.

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